

AD-A110 309

INDIANA UNIV AT BLOOMINGTON DEPT OF PHYSICS
OPTICAL PUMPING STUDIES OF COLLISIONAL INTERACTIONS.(U)
NOV 81 F A FRANZ

F/G 20/6

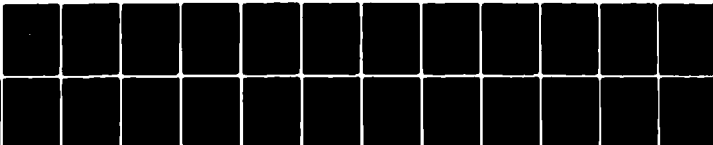
AFOSR-79-0031

UNCLASSIFIED

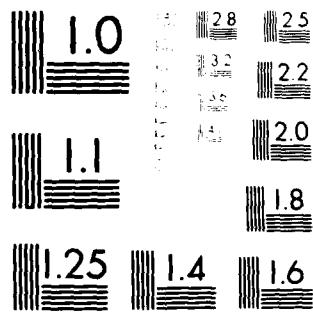
AFOSR-TR-81-0897

NL

105
411
1000000



END
DATE
FILMED
102-82
DTIC



MICROCOPY RESOLUTION TEST CHART
NATIONAL BUREAU OF STANDARDS-1963-A

AFOSR-TR. 79-0031

LEVEL

13

24 NOV 1982

Final Scientific Report

"Optical Pumping Studies of Collisional Interactions"

Supported by

Air Force Office of Scientific Research

AFOSR [REDACTED] 79-0031

Principal Investigator

F.A. Franz

Department of Physics

Indiana University

Bloomington, Indiana 47405

DTIC
FEB 1 1982
H

AD A110309

DTIC FILE COPY

82 01 8036

Approved for public release;
distribution unlimited.

400-112

REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER AFOSR-TR- 21-0897	2. GOVT ACCESSION NO. AD-A110 309	3. RECIPIENT'S CATALOG NUMBER
4. TITLE (and Subtitle) OPTICAL PUMPING STUDIES OF COLLISIONAL INTERACTIONS		5. TYPE OF REPORT & PERIOD COVERED Final 1 Jan 79 - 30 Jun 81
		6. PERFORMING ORG. REPORT NUMBER
7. AUTHOR(s) F.A. Franz		8. CONTRACT OR GRANT NUMBER(s) AFOSR 79-0031
9. PERFORMING ORGANIZATION NAME AND ADDRESS Indiana University Bloomington, Indiana 47405		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS 61102F 2301/A4
11. CONTROLLING OFFICE NAME AND ADDRESS AFOSR/NP Building 410 Bolling AFB, DC 20332		12. REPORT DATE November 23, 1981
		13. N 25 PAGES
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)		15. SECURITY CLASS. of this report Unclassified
		15a. DECLASSIFICATION/DOWNGRADING SCHEDULE
16. DISTRIBUTION STATEMENT (of this Report) Approved for Public Release: Distribution Unlimited		
17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)		
18. SUPPLEMENTARY NOTES		
19. KEY WORDS (Continue on reverse side if necessary and identify by block number) Optical pumping, Spin Relaxation, Atomic Collisions, Alkali Metals, Cs, Rb, K, Nuclear Spin, Depolarization, Selection Rules, He, Ne, Ar. Wall relaxation, Masers, magnetic polarization, orientation, quasi bound molecules, van der Waals molecules, hyperfine interaction, gyroscopes.		
20. ABSTRACT (Continue on reverse side if necessary and identify by block number) A final scientific report is given on a project involving application of optical pumping techniques to studies of atomic collisions. Magnetic depolarization, nuclear spin efforts, collisional selection rules, surface interactions, and studies of sudden and quasi molecular collisions between alkali atoms and noble gas atoms are particularly emphasized.		

I. Resume of Results of Research

The primary goal in our AFOSR sponsored research has been the application of optical pumping techniques to the study of certain kinds of atomic collisional interactions. We have been particularly interested in studying various modes of relaxation of electronic and nuclear spin polarizations induced in collisions of magnetically polarized atoms with foreign (buffer gas) atoms. Such studies have practical significance in providing insight into the physical phenomena which govern the operation and performance of magnetometers, masers, nuclear gyroscopes, and secondary frequency standards. They are of fundamental interest through the unique information they provide on interatomic interactions.¹

A. Optical Pumping Transients in Weak Magnetic Fields

1. Background

Electronic and nuclear spin polarizations are produced in optical pumping through the absorption and re-emission of circularly polarized resonance radiation. These polarizations correspond to artificially created imbalances in the relative populations of the Zeeman sublevels of atomic ground states. The degree of polarization attained and the exact distribution of relative populations produced throughout the Zeeman sublevels depends upon the pumping process itself and upon a variety of forms of collisional relaxation, both in the ground and excited atomic states. In all such processes the hyperfine interaction between the electronic magnetic moment and the nuclear spin can play an extremely important and complicating role.²⁻⁵

AIR FORCE OFFICE OF SCIENTIFIC RESEARCH (AFOSR)
NOTES
SERIAL
AND
DATE
MAITHEE, J. J.
Chief, Technical Information Division

One can observe the evolution of a system undergoing optical pumping by monitoring the intensity of light scattered or transmitted by the optical pumping cell. Under the appropriate conditions the time constants of the transient pumping signals directly reflect the characteristics of the various relaxation processes. In earlier research we brought together all relevant knowledge of the mechanics of the pumping and relaxation processes and obtained analytic solutions describing pumping transients in alkali metal vapors, subject to the condition that the pumping rate is weak compared to relaxation rates.^{8,9} We showed within well established approximations how collisions of pumped atoms with the walls of the experimental cell, simple binary alkali-atom-buffer-gas atom collisions, and alkali-alkali spin exchange collisions all contribute to determining the shape of these transients. We included rigorous treatments of the effect of the hyperfine interaction on all pumping and collisional relaxation processes. In low magnetic fields the pumping transients of the electronic spin polarization are generally represented by the sum of two exponentials, the rate constants of which depend wholly upon relaxation processes in the ground state, with the relative contributions of the two rates to the production of the ground state electronic spin polarization ($\langle S_z \rangle_g$), being determined mainly by the degree of collisional relaxation which occurs during the excited state lifetime. The comprehensive analytic expressions we derived involved all known relaxation processes: they offered new and dependable ways to measure cross sections for electronic and nuclear spin relaxation in sudden binary collisions between alkali metal atoms and noble gas atoms both in the ground and excited states of the alkali metal atoms. They led to the following simple form of the optical pumping transient subject to weak D_1 + optical pumping:

Distribution For Distribution/Availability Codes Avail and/or Special	<input checked="checked" type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/>
A	

$$\langle S_z \rangle_g = D_1(1 - \exp(-Z_1 t)) + D_2(1 - \exp(-Z_2 t)), \quad (1)$$

where Z_1 and Z_2 are functions of all relaxation and pumping processes occurring in the ground state, and D_1 and D_2 depend in addition upon the rate of collisional relaxation in the $^2P_{1/2}$ state. The double exponential nature of the transient is a consequence of nuclear spin mechanics.

Application of Eq. 1 forms the basis for the two experiments described below.

2. The effect of the formation and destruction of quasi bound alkali-metal-atom-light-noble-gas atom van der Waals molecules on electronic and nuclear spin relaxation: relaxation of ^{39}K and ^{41}K in He and Ne.

The best known cause of electronic spin relaxation is sudden binary collisions of alkali metal atoms with noble gas atoms is the effective spin orbit interaction, $\gamma(\vec{S} \cdot \vec{N})$, induced between S , the spin of the valence electron of the alkali metal atom, and N , the angular momentum of the alkali metal atom and the noble gas atom around each other.^{2,6,7} The $\gamma(\vec{S} \cdot \vec{N})$ interaction is important both in sudden binary collisions and in molecule-forming or "sticky" collisions.¹¹ In the case of Rb in moderate pressures of He and Ne, however, the dominant mode of relaxation has been shown to be an anomalous process, attributed to the modification of the Rb hyperfine interaction, $\delta a(S \cdot I)$ in the formation of Rb-He and Rb-Ne van der Waals molecules.⁹

In research supported by this grant we used low magnetic field white light optical pumping to measure the electronic and nuclear spin relaxation of the $4^2S_{1/2}$ state of K. Our primary interest lay in determining the degree to which anomalous relaxation might exist in an alkali metal with a significantly smaller hyperfine interaction than Rb. A secondary interest was that spin

relaxation rates for potassium had been measured previously only in He, and had been evaluated at a time when the influence of nuclear spin mechanics on optical pumping transients was not fully appreciated. There was the promise, therefore, also of gaining new information on the $\gamma (\vec{S} \cdot \vec{N})$ cross sections for K.

We summarize below the relaxation mechanisms which are expected to make contribution to the relaxation rates, Z_1 and Z_2 , in the present experiment.

- a. $\gamma (\vec{S} \cdot \vec{N})$ relaxation in sudden binary collisions of ground state K atoms with He or Ne atoms should make a full contribution, R , to Z_2 , but only $R/8$ to Z_1 . The difference in the two contributions is a consequence of nucleon spin mechanics. The $\gamma (\vec{S} \cdot \vec{N})$ relaxation rate should be proportional to the pressure of the buffer gas.
- b. Relaxation resulting from diffusion of K to the walls of the cell, R' , makes equal contributions to Z_2 and to Z_1 . This relaxation rate is inversely proportional to buffer gas pressure.
- c. K-K spin exchange, R_s , makes a strong contribution to Z_2 , but essentially no contribution to Z_1 . R_s is independent of buffer gas pressure.
- d. Anomalous relaxation, R^* , in the K ground state should make a full contribution to Z_2 , but no contribution to Z_1 , if R_s is of the form $\delta a (\vec{S} \cdot \vec{I})$.

In Fig. 1 we display measurements of the "slow" and "fast" relaxation rates, Z_1 and Z_2 , for optically pumped ^{39}K in He over the range of He pressures from 30 Torr to 550 Torr. R , the binary $\gamma(\vec{S} \cdot \vec{N})$ relaxation rate, already small for K-He, makes even smaller contributions to Z_1 , being reduced by the nuclear spin "slowing factor", $1/8$. Z_1 therefore reflects mainly the wall relaxation rate, R' , and the pumping rate, $A/3$. Z_2 reflects R' and $A/3$, and in addition contains full contributions from R and R_s . A "hump" in the Z_2 data between 100 Torr and 300 Torr is prominent: this maximum is similar to that observed for Z_2 of Rb in He, and has the properties of anomalous relaxation, R^* that is, a strong contribution to Z_2 , but no discernable contribution to Z_1 .

In Fig. 2 we display measurements of Z_1 and Z_2 for ^{41}K in Ne. As is the case for K in He, there is a pronounced anomalous effect in the Z_2 data.

Analysis of the data in Figs. 1 and 2, and subsidiary results lead to experimental determinations of the diffusion coefficients of K in He and K in Ne, and to that cross sections for the distinction of spin polarization via the $\gamma(\vec{S} \cdot \vec{N})$ interaction. Figs. 1 and 2 also make clear, however that anomalous relaxation plays important roles in K-He and K-Ne. For K-He, R^* at 200 Torr is about 3 sec^{-1} , whereas $R(\gamma(\vec{S} \cdot \vec{N}))$ is about 0.66 sec^{-1} : at that pressure of He R^* is about 4.5 times greater than R . Similarly, at 100 Torr of Ne, R^* is about 3 sec^{-1} , whereas $R(\gamma(\vec{S} \cdot \vec{N}))$ is about 4 sec^{-1} : the contributions from the two relaxation processes are comparable. R^* is considerably less significant in K than in Rb, however, where maximum values of R^* are about five times greater, and where the dominance of R^* over $R(\gamma(\vec{S} \cdot \vec{N}))$ can be by as much as a factor of forty.

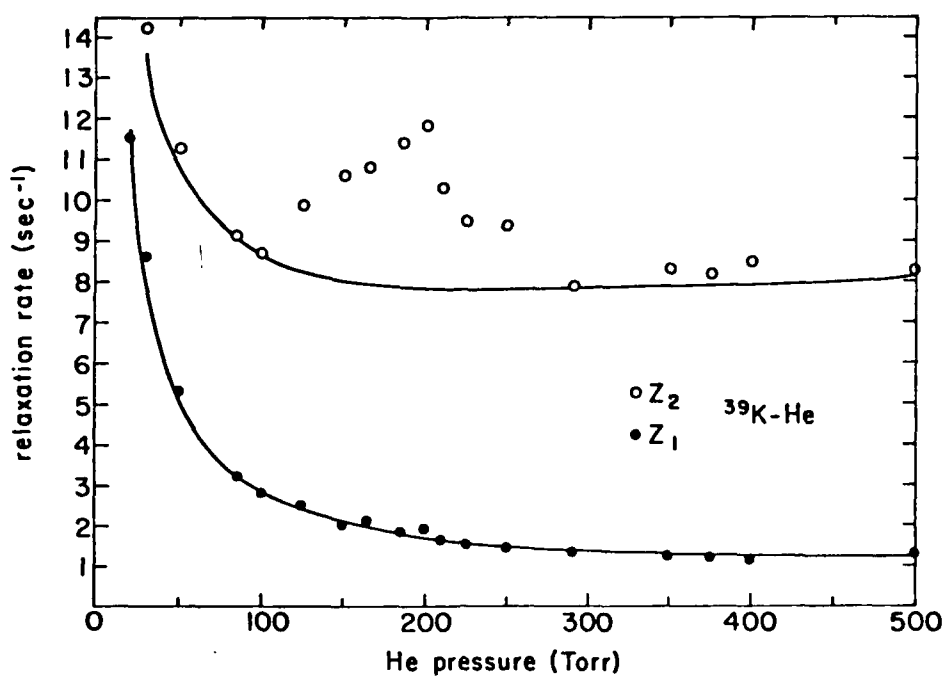


Fig. 1. Measured relaxation rates, Z_1 and Z_2 , for ^{39}K in He. Each data point represents the average of three to five measurements. The solid curves are fits to the data, ignoring anomalous relaxation.

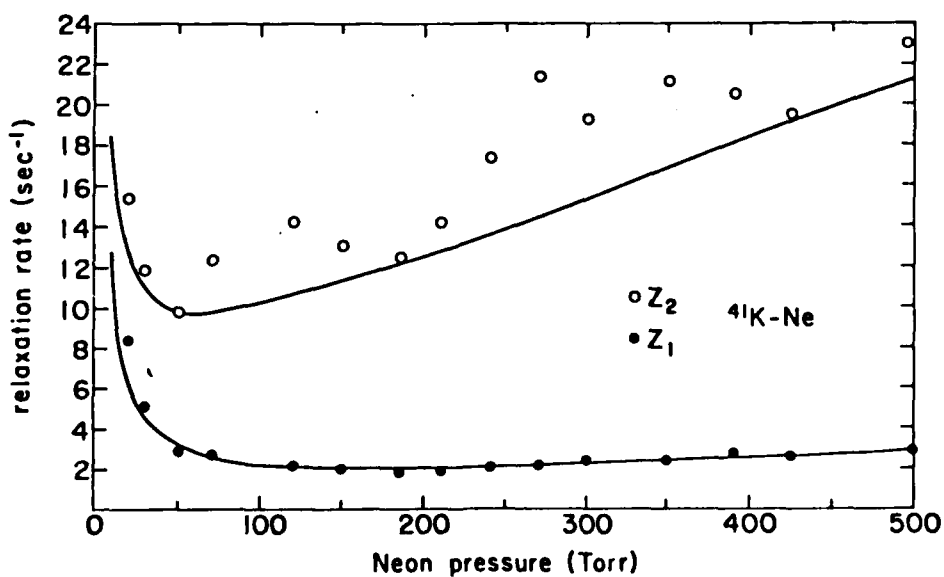


Fig. 2. Similar to Fig. 1, but for ^{41}K in Ne.

In earlier research we attributed the cause of anomalous relaxation to the $\delta a(S \cdot I)$ interaction induced in the formation of alkali-noble-gas van der Waals molecules.⁹ The K results are compatible with that interpretation. The ideas underlying this explanation, briefly, are these. Sudden collisions between atoms last a time of the order of that required for the two atoms to "fly by" each other, in other words, about 10^{-13} sec at 300°K. If, however, an alkali atom collides simultaneously with two noble gas atoms, a weakly bound alkali-atom-noble-gas-atom van der Waals molecule may be formed with the third atom carrying off the appropriate amount of excess energy.¹² Such a molecule generally lives only until hit by another buffer gas atom. At pressures of the order of several Torr this time may be of the order of 10^{-7} sec, very long compared to the duration of a fly-by collision. The significantly longer duration of perturbation in the molecular case can result in much greater probability of relaxation. Thus even though events of molecular formation may be relatively rare, they may still have great influence ultimately on spin relaxation rates.

Still another type of alkali-noble-gas collision is possible. The effective potential for an alkali-atom-noble-gas-atom pair is the combination of the van der Waals potential and the centrifugal barrier potential ($V(r)$). True bound states formed in three body collisions are those with energies less than zero on the $V(r)$ scale. There are the possibilities of quasi-bound states, however, with energies greater than zero, but less than the height of the barrier potential. Such quasi-bound molecules can be formed in the same way that normal bound molecules are formed, i.e., in three body collisions. They also can be formed

in two body collisions, i.e. an alkali-atom-buffer-gas-atom pair of the appropriate total energy can "tunnel" through the barrier to form a quasi-bound state. Quasi-bound complexes should behave much as the bound states except for the fact that they have a natural dissociation (tunneling) rate in addition to the collisional destruction rate.

Contributions of molecular formation to the rate of spin relaxation of Rb in the heavy noble gases Kr and Xe were demonstrated and studied extensively by Bouchiat and co-workers.¹² They showed that the relaxation mechanism was the same as that present in binary collisions, namely $\gamma(\vec{S} \cdot \vec{N})$, acting for a much longer time. In the case of $\gamma(\vec{S} \cdot \vec{N})$ relaxation via molecular formation, theory predicts strong contributions of the anomalous relaxation rate to both exponential terms in the $\langle S_z \rangle_g$ relaxation transient. In our work, however, we find a strong contribution to the "fast" rate constant, but an essentially zero contribution to the "slow" rate constant. There is no way for the $\gamma(\vec{S} \cdot \vec{N})$ interaction in molecular formation to fit the observed experimental facts. The K-He and K-Ne results show this effect quite dramatically.

An approximate treatment of anomalous relaxation via the $\delta a(\vec{S} \cdot \vec{I})$ interaction in van der Waals molecular formation yields Eq. 2:⁹

$$R^*(\delta a(\vec{S} \cdot \vec{I})) \approx \frac{(\text{Const}) \tau_{e2}^2 T_f^{-1} \frac{\omega_s^2}{\Delta W}}{1 + (\Delta W)^2 \tau_{e2}^2}, \quad (2)$$

where $(\tau_{e2})^{-1}$ is the rate of breakup of molecular complexes, T_f^{-1} is their formation rate, W is the angular hyperfine frequency, and ω_s is the electronic Larmor frequency in the external magnetic field (H_0).

The R^* data both for K-He and for K-Ne display marked maxima: the K-Ne data in fact appear to display two maxima. The most likely cause of maxima in R^* are contributions to R^* arising from the formation of van der Waals complexes in two body rather than three body collisions. At low pressure the R^* relaxation rate is effectively proportional to the formation rate of complexes, thus proportional to p in the two body case and to p^2 in the three body case. At higher pressures the destruction rate shortens the value of the correlation time τ_{e2} ; R^* should approach zero in the two body case, but approach a constant value in the three body case. The accuracy of our data for R^* and the accuracy of our determination of $\sigma(\vec{S} \cdot \vec{N})$ do not permit us to say with certainty what the equilibrium value of R^* is at high pressure, either in He or in Ne. Fig. 1 suggests, for example, that $R^*(K - He)$ at high He pressures is about 0.5 sec^{-1} , but a value of zero cannot be excluded. Similarly, there appears to be a second maximum of $R^*(K - Ne)$ in Fig. 2, but this apparent peak could instead, within experimental uncertainty, be a rise of R^* to a constant value.

A pressure dependence of R^* for K-Ne similar to that observed could result if the breakup cross section for quasibound K-Ne molecules was substantially different from that for bound K-Ne molecules. In the bound complex case, the break-up process must add some energy to the molecular system in order to free the alkali and noble gas atoms. For the quasi-bound states, however, a distortion of the internuclear potential either to a degree which actually frees the alkali and noble gas atoms or simply increases the tunneling probability is sufficient to break-up the van der Waals molecule. Such a process is

represented by a substantially larger cross section than the bound state break-up cross section. The single peak in the K-He R^* rate may imply that only the quasi-bound states are involved. This is compatible with calculations based on the Baylis potential parameters,¹⁶ which for the case of K-He indicate that there is a single quasi-bound state with a six-fold degeneracy and one bound state with a two-fold degeneracy. In this case the quasi-bound states would dominate and the R^* rate might exhibit only one peak as a function of pressure. Further work on these matters is planned for the future.

3. Relative transition probabilities in atomic de-excitation by collisional quenching.

Radiationless de-excitation of excited atoms produced by quenching collisions with molecules has been widely studied for many years.¹³⁻¹⁷ Generally unknown, however, are the explicit paths that quenched atoms follow from the excited state to the ground state. With what relative probabilities does an atom in a particular Zeeman sublevel of an atomic excited state undergo transition via quenching to various sublevels of the ground state? Are electronic and nuclear spin polarizations preserved in the process? Classical measurements of the polarization of resonance radiation yield no information on these questions: quenched atoms yield no photons for analysis. The path that a quenched atom follows from the excited state to the ground state is in effect an invisible one.

The generation of nuclear and electronic spin polarizations in optical pumping, however, is sensitive to the detailed mode of repopulation of the ground state from the optically pumped excited state. With AFOSR sponsorship

we have developed ways in which we have used optical pumping experiments to measure relative quenching probabilities.¹⁸ The influence of quenching upon optical pumping is of practical as well as fundamental interest since relative probabilities for quenching affect the efficiency of optically pumped devices such as magnetometers, masers, and frequency standards.¹

In an earlier paper we had developed a theory which allowed relative probabilities for quenching transitions between $|F', m_{F'}\rangle$ sublevels of a $^2P_{1/2}$ excited state and the $|F, m_F\rangle$ sublevels of the $^2S_{1/2}$ ground state of a heavy alkali metal atom to be expressed in terms of nuclear spin independent transition probabilities connecting $|J', m_{J'}\rangle$ and $|S, m_S\rangle$ sublevels.¹⁸ The theory was applied to analyses of optical pumping transients of Cs in N_2 under conditions where the lifetime against quenching was long compared to the hyperfine period. In work supported by the present grant we examined the influence of quenching upon the optical pumping of ^{85}Rb and ^{87}Rb . First, we extended the calculations of the previous work to the cases of nuclear spin $I = 3/2$ and $I = 5/2$, relevant for ^{85}Rb and ^{87}Rb . Tables of the calculated relative probabilities and of repopulation rates are provided in a paper to be published.

The hyperfine interactions in the $^2P_{1/2}$ states of ^{85}Rb and ^{87}Rb are small enough, however, that the original theory applies only at collision rates corresponding to N_2 pressures of a few Torr or less. We therefore provided a new calculation which includes the situation where the quenching rate is large compared to the hyperfine frequency of the excited state. Since calculations of relative probabilities for quenching treat nuclear spin effects in a separable way, experimental results for the two isotopes provide an important test of the theory.

We used these results to show how quenching collisions affect the efficiency of the optical pumping process.

Experimental results are shown in Fig. 3. The ordinate of the graph, C_1/B_1 , represents our experimental determination of the ratio of the pumping plus repopulation rates of $\langle I_z \rangle_g$ and $\langle S_z \rangle_g$, respectively. The solid curve is the result of a fit of our theoretical calculation to the data. From this fit we determine both the relative probabilities for de-excitation via quenching and the cross section for collisional destruction of orientation in the $5^2P_{1/2}$ excited state of Rb. It may be interesting to note that according to the original theory (strong hyperfine structure, moderate quenching rates) C_1/B_1 would be predicted to be essentially independent of N_2 pressure.

Conservation of electronic spin polarization in quenching collisions

It has been argued that if the quenching interaction is sudden and of short duration compared to the $\vec{L} \cdot \vec{S}$ period, then de-excitation through quenching should occur with the orientation of the electronic spin left unchanged: the relative quenching probabilities would be subject to the selection rule $\Delta m_s = 0$. Our results are consistent with this prediction and thus suggest that electronic spin polarization is conserved in the quenching of the $5^2P_{1/2}$ state of Rb by N_2 , and that the duration of the quenching interaction is short compared to the fine structure period (1.4×10^{-13} sec.).

Influence of quenching on efficiency of optical pumping

Our experimental and theoretical results provide insight into the influence of quenching interactions on the efficiency of the optical pumping process. Under some conditions quenching improves efficiency; under others, it diminishes it.

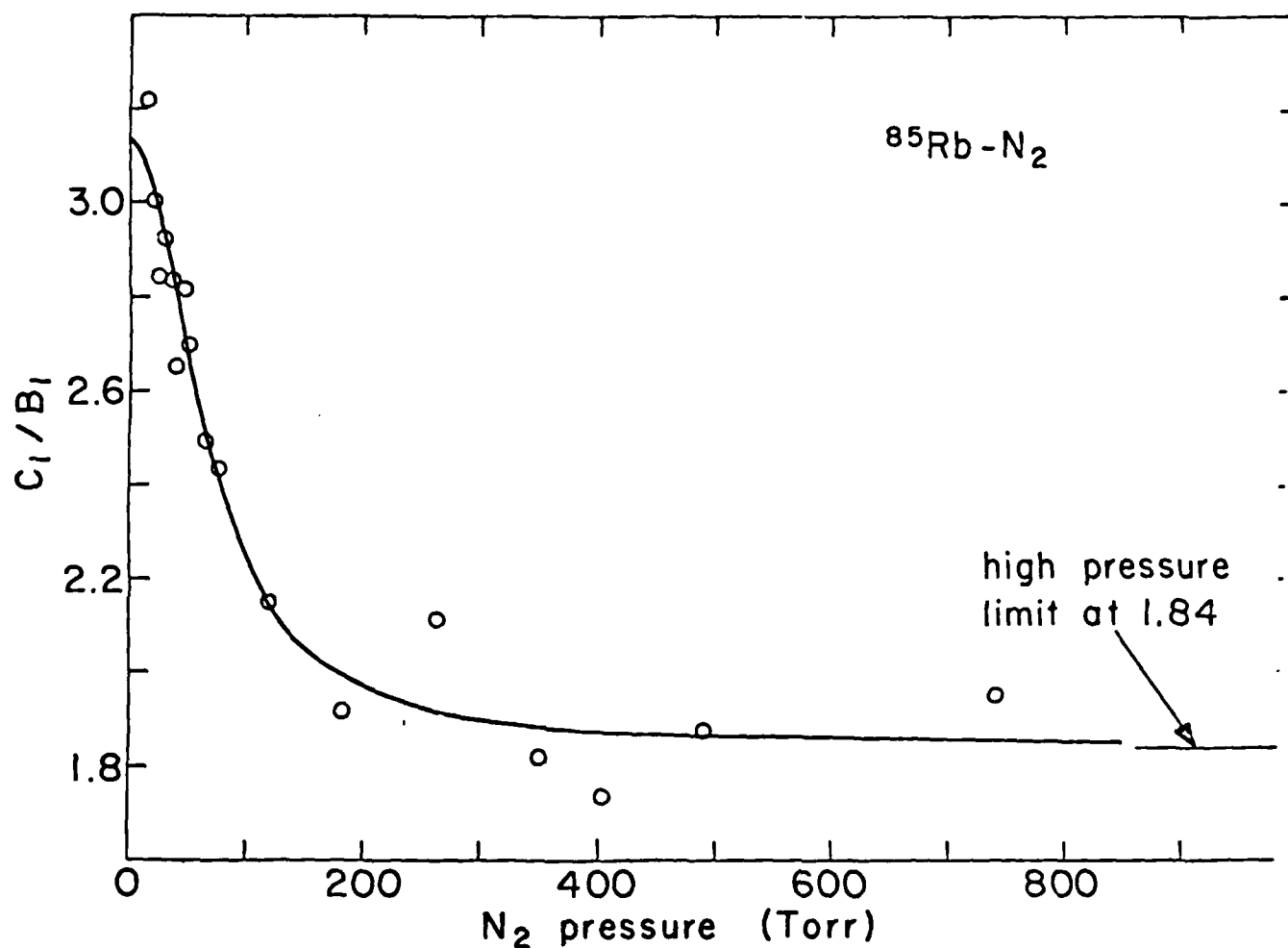


Fig. 3 Experimentally determined values of C_1/B_1 for optically pumped ^{85}Rb as a function of pressure of N_2 . The solid curve is a theoretical fit to the data. The pressure dependence of C_1/B_1 is due primarily to the effect of the hyperfine interaction and the shortening of the lifetime against quenching. The original theory predicts no dependence of C_1/B_1 upon N_2 pressures above a few Torr.

Quenching always provides important enhancement of pumping efficiency through removal of fluorescent photons which otherwise would be emitted as a by-product of the pumping process. Reabsorption of such photons by pumped atoms would act as a relaxation mechanism on ground state electronic and nuclear spin polarization, an effect particularly important at alkali metal densities high enough to be subject to radiation trapping. This beneficial effect is always present: its contribution is not treated by considerations in our work. Our results make clear, however, why a little N_2 may enhance efficiency, but a lot may harm it. From our work we find that at high N_2 pressures C_1 drops to about half of its value at low pressure, with a corresponding decrease in equilibrium spin polarization. The degradation of pumping efficiency at high N_2 pressures is a direct result of the shortened lifetime of the quenched excited state. Optical pumping devices often contain a relatively high pressure of buffer gas to retard relaxation at the walls of the cell. The present results indicate why optimal pumping efficiency should be obtained if a small partial pressure (a few Torr) of N_2 is present to quench the fluorescent photons, but the dominant contribution to buffering is provided by a non-quenching gas. There are several important paths related to this effect which should be followed in future research.

B. COLLISIONAL INTERACTIONS IN VERY HIGH MAGNETIC FIELDS

1. Background

In earlier AFOSR sponsored research we performed an experiment involving the optical pumping of cesium in magnetic fields ranging up to 100 kG. This work constituted the first study of collisional relaxation of electronic spin polarization in very high magnetic fields.¹⁹ There were some intriguing discoveries. First, in strong magnetic fields (of the order of 20 kG or greater), one should be able to measure electron spin relaxation fully decoupled from effects of the hyperfine interaction. We verified this in Cs-Ar, where we measured a relaxation cross section of $100 \times 10^{-23} \text{ Å}^2$. The high field value agrees well with our low magnetic field measurement of the nuclear spin independent cross section ($108 \times 10^{-23} \text{ Å}^2$) and the measurement of $104 \times 10^{-23} \text{ Å}^2$ for the cross section for relaxation of $\langle \vec{S} \cdot \vec{I} \rangle$ measured by Beverini et. al. According to theory all these cross sections should be equal -- they are, in fact, theoretically the same entity.

The results in Cs-Ne and Cs-He, however, were puzzling. Rather than finding a cross section equal to the nuclear spin independent cross section, we found equivalent high field cross sections to be approximately 2.5 times larger than the low field values. The full explanation of this effect is given in ref. 19. Summarizing briefly, if the collisional spin orbit interaction, $\gamma(\vec{S} \cdot \vec{N})$, were the only interaction present between alkali metal atoms and buffer gas atoms, one should measure equal relaxation cross sections at low and at moderately high magnetic field. However, collisions also perturb the alkali metal atom hyperfine interaction, $a(\vec{S} \cdot \vec{I})$, giving rise to the well known pressure shift of hyperfine

frequencies in optically pumped masers and frequency standards. The $a(\vec{S} \cdot \vec{I})$ interaction also contributes to relaxation rates at low or intermediate magnetic fields according to the following equation:^{9,18}

$$R_2(\delta a(\vec{S} \cdot \vec{I})) \cong \frac{8\pi^2}{3} (\delta u)^2 \frac{(n_o \sigma_c v_{rel}/p_o)P}{(1 + \omega_o^2 \tau_{e2}^2)} \frac{\omega_s^2}{\Delta W}, \quad (2)$$

where δu is the measured hyperfine frequency shift in Hz/Torr, σ_c is an average cross section for the $\delta a(\vec{S} \cdot \vec{I})$ interaction in binary collisions, p is the buffer gas pressure in Torr, and τ_o , τ_{e2} , ω_s , and ΔW have been defined in section I above. At low magnetic fields the factor $(\omega_s/\Delta W)^2$ is small, generally making R_2 negligible compared to other relaxation rates. At fields above a few kG, however, the correct expression for R_2 is obtained by letting $(\omega_s/\Delta W) = 1$, that is

$$R_2(\delta a(\vec{S} \cdot \vec{I})) (\text{High } H_o) \cong \frac{8\pi^2}{3} (\delta u)^2 \frac{(n_o \sigma_c v_{rel}/p)P}{1 + \omega_o^2 \tau_{e2}^2}. \quad (3)$$

Calculations show that for Cs-Ne and Cs-He the contributions of the $\delta a(\vec{S} \cdot \vec{I})$ interaction to relaxation rates in fact are greater than those for $\delta(\vec{S} \cdot \vec{N})$. In research supported by the present grant we extended measurements to Rb and found truly intriguing results.

1. Measurement of the relaxation of S_z in the $^2S_{1/2}$ ground states of Rb⁸⁵ and Rb⁸⁷ at high magnetic field. The only difference in Eq. 3 for Rb⁸⁵ and Rb⁸⁷ at high magnetic field lies in the factor (δu) : (δu) for Rb⁸⁷ is approximately 2.25 times larger than that for Rb⁸⁵. Measurement of the relaxation rates of the separated isotopes at high magnetic field therefore is important for the confirmation of the $\delta a(\vec{S} \cdot \vec{I})$ interaction's contributions to

relaxation rates: $\delta a(\vec{S} \cdot \vec{I})$ relaxation rates for ^{87}Rb should be about five times greater than those for ^{85}Rb .

A full description of our experiment has appeared in the June, 1981 issue of Physical Review A.¹⁹ Summarizing briefly, the experimental results confirmed our theoretical predictions. In Figs. 4 and 5 we show the actual relaxation rates measured for ^{85}Rb and ^{87}Rb in He and Ne. In Figs. 6 and 7 we show "reduced data" representing that part of the relaxation rate contributed by the $\delta a(\vec{S} \cdot \vec{I})$ interaction. The $\delta a(\vec{S} \cdot \vec{I})$ binary relaxation rate indeed turns out to be roughly 5 times greater for ^{87}Rb as for ^{85}Rb . From the data we also determined cross sections for the $\delta a(\vec{S} \cdot \vec{I})$ interaction in the Rb-He and Rb-Ne collisions.

The experiment was the first conclusive demonstration of the importance of the $\delta a(\vec{S} \cdot \vec{I})$ interaction in collisional relaxation: The data also contains important information on the role of the $\delta a(\vec{S} \cdot \vec{I})$ interaction on relaxation induced by the formation of Rb-He and Rb-Ne van der Waals molecules. We hope to pursue that course of inquiry in the future.

2. Relative probabilities for intramultiplet mixing

In our grant proposal I pointed out that in recent years there has been intensive and widespread study of collisionally induced transitions between atomic multiplets, especially between the various $^2P_{1/2}$ and $^2P_{3/2}$ states of alkali metal atoms.²⁰

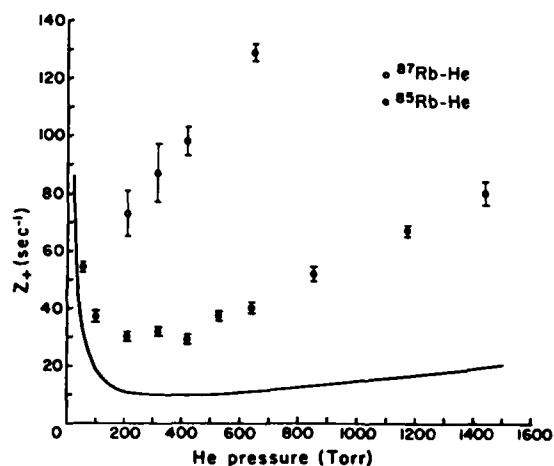


FIG. 4. Measurements of the "fast" relaxation rate Z_+ for ^{85}Rb and ^{87}Rb in He. Uncertainties are ± 1 standard deviation. The solid curve represents an extrapolation from low-field measurements ignoring the $\delta a(\vec{S} \cdot \vec{I})$ interaction.

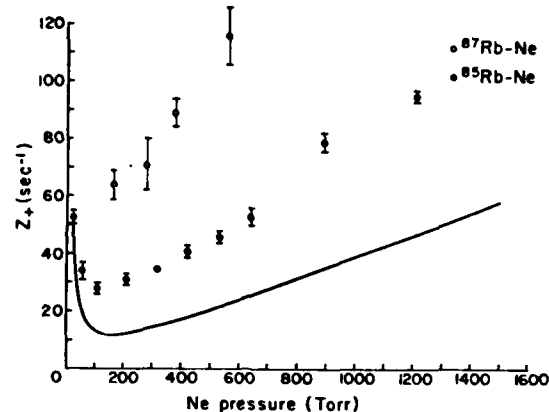


FIG. 5. Measurements of the fast relaxation rate Z_+ for ^{85}Rb and ^{87}Rb in Ne. Uncertainties are ± 1 standard deviation. The solid curve represents an extrapolation from low-field measurements ignoring the $\delta a(\vec{S} \cdot \vec{I})$ interaction.

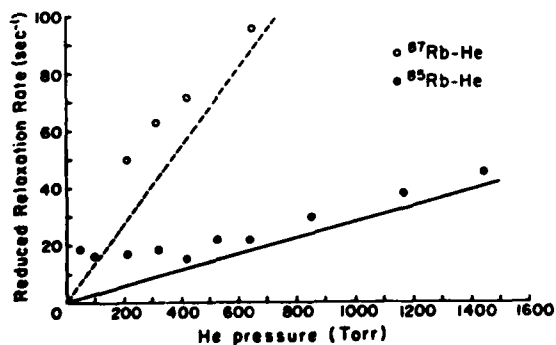


FIG. 6. "Reduced data" for ^{85}Rb and ^{87}Rb relaxation in He at 40 kG.

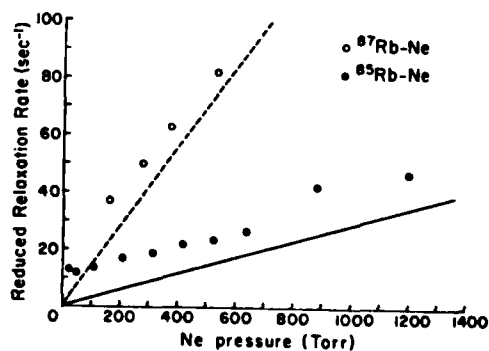


FIG. 7. Reduced data for ^{85}Rb and ^{87}Rb relaxation in Ne at 40 kG.

It is of particular interest to measure experimentally the relative probabilities connecting Zeeman sublevels in collisionally induced intermultiplet transitions.²¹ Such measurements provide tests of the various theoretical descriptions of the collisional transfer interaction. The only way that full information of this sort can be obtained is to make a selective excitation of atoms in a particular Zeeman sublevel, and observe resonance fluorescence from other sublevels populated by collisional transfer. It requires energetic resolution of the individual components of the various spectral lines and decoupling of nuclear spin effects. Our high magnetic field facility is ideally suited for such experiments -- Indiana is, in fact, one of the few places in the world where this work can be done.

In research supported by this grant we carried out measurement and analysis of collisional transition probabilities connecting all (J, m_J) sublevels of the $4^2P_{1/2}$ and $4^2P_{3/2}$ states of potassium colliding with He, Ne, or Kr. The work was done at 22 kG and 131 kG, and constitutes the first measurement of these parameters.

Our results can be combined in various ways to compare with less complete results of others. For example, our results indicate cross sections for the destruction of orientation and alignment in the $^2P_{3/2}$ state which are consistent with earlier measurements of these parameters by Berdowski, Shiner, and Krause.²² Our results confirm a previously observed dependence of the cross section for relaxation within the $^2P_{1/2}$ state upon magnetic field,²³ but cast into doubt some earlier measurements of $^2P_{1/2}$ state relaxation made by Berdowski and

Krause.²⁴ Likewise, our results for $2P_{1/2} \leftrightarrow 2P_{3/2}$ transitions adjudicate between conflicting results of several researchers. Perhaps most important, however, a calculation of the various relaxation cross section made by Pascale and Perrin,²⁵ based on potential parameters of Baylis,²⁶ are fully consistent with all of our cross sections, an impressive confirmation of both of the potential and of the modes of calculation. More complete discussion appears in the article submitted for publication.

II. Journal Articles Published or Submitted Reporting AFOSR Sponsored Research Completed During the Grant Period.

1. "Spin relaxation of rubidium atoms induced by collisional modification of the Rb hyperfine interaction", F.A. Franz and A. Sieradzan, Phys. Rev. A 23, 2841 (1981). Spin relaxation of Rb atoms induced by collisional modification of the hyperfine interaction was measured for ^{85}Rb and ^{87}Rb in He, Ne, and Ar buffer gases at high magnetic field. The relaxation rates for ^{87}Rb were shown to be approximately 4.9 times larger than those for ^{85}Rb , in accord with theory: Cross sections for the $\delta a(\vec{S} \cdot \vec{I})$ binary collisional interaction were measured. Anomalous relaxation, attributed to the $\delta a(\vec{S} \cdot \vec{I})$ interaction in Rb-noble-gas van der Waals molecules was found to be present at high field to at least the same degree as observed at low magnetic field.
2. "Cross sections for $J, m_J \rightarrow J', m_{J'}$ transitions within the 4^2P states of potassium induced by collisions with He, Ne and Kr," R. Boggy and F.A. Franz, submitted to Phys. Rev. A(1981). Selective laser excitation and monitoring of fluorescence of individual (J, m_J) Zeeman sublevels of the $4^2P_{1/2}$ and $4^2P_{3/2}$ states of K at 22 kG and 131 kG were used to measure $(J, m_J \rightarrow J', m_{J'})$ transition cross sections for K colliding with He, Ne, and Kr. The results were translated into cross sections for the destruction of orientation, alignment, and octapolar orientation within the $4^2P_{3/2}$ state and or orientation within the $4^2P_{1/2}$ state. The experiment also provided measurements of intra-

multiplet ($2P_{1/2} \rightarrow 2P_{3/2}$) transitions. The results agreed well with theoretical calculations based on the interatomic potential of Baylis.

3. Electronic spin relaxation of the $4^2S_{1/2}$ state of Potassium induced by K-He and K-Ne collisions," by F.A. Franz and C. Volk. Electronic and nuclear spin relaxation of optically pumped K atoms was measured in the presence of He and Ne buffer gases. Nuclear spin independent cross sections for the destruction of electronic spin polarization in the $4^2S_{1/2}$ ground state of K were measured, as were diffusion coefficients for K in He and Ne. The relaxation of K was shown to be strongly affected by anomalous relaxation attributed to the modification of the K hyperfine interaction induced by the formation of K-He and K-Ne van der Waals molecular complexes.

4. "Quenching, depolarization and transfer of spin polarization in Rb-N₂ collisions", by A. Sieradzian and F.A. Franz, submitted to Phys. Rev. A (1981). The transfer of electronic and nuclear spin polarization from the $5^2P_{1/2}$ state of Rb to the $5^2S_{1/2}$ state caused by quenching collisions of Rb atoms with N₂ molecules was studied. The dependence of such polarization transfer upon the strength of the hyperfine interaction in the excited state was evaluated; and nucleon spin independent relative probabilities connecting the J, m_J sublevels of the $5^2P_{1/2}$ state with the S, m_S sublevels of the $5^2S_{1/2}$ ground state were evaluated. The cross section for the destruction of orientation in the $5^2P_{1/2}$ state also was measured. The results showed how quenching can cut to improve optical pumping efficiency.

REFERENCES

1. For extensive reviews of many theoretical and experimental aspects of optical pumping relevant to the present work, see W. Happer, Rev. Mod. Phys. 44, 169 (1972), and "Optical Pumping," by L.C. Balling, in Advances in Quantum Electronics 3, ed. by D.W. Goodwin Academic Press, New York, 1975.
2. M.A. Bouchiat J. Phys. (Paris) 24, 370 (1963); 24, 611 (1963); M.A. Bouchiat and J. Brossel, Phys. Rev. 147, 41 (1966). F. Masnou-Seeuws and M.A. Bouchiat, J. Phys. 28, 406 (1967).
3. F.A. Franz and J.R. Franz, Phys. Rev. 148, 82 (1966).
4. J.F. Papp and F.A. Franz, Phys. Rev. A 5, 1763 (1972).
5. F.A. Franz, Zeitschrift fur Physik 262, 105 (1973).
6. R.A. Bernheim, J. Chem. Phys. 36, 135 (1962).
7. R.M. Herman, Phys. Rev. 136, A1576 (1964).
8. F.A. Franz and C.E. Sooriamoorthi, Phys. Rev. A8, 2390 (1973); A10, 126 (1974).
9. F.A. Franz and C. Volk, Phys. Rev. Letters 35, 1704 (1975); Phys. Rev. A14, 1711 (1976); Phys. Rev. A18, 599 (1978).
10. C.C. Bouchiat, M.A. Bouchiat, and L.C.L. Pottier, Phys. Rev. 181, 146 (1979).
11. See, for example, C.C. Bouchiat and M.A. Bouchiat, Phys. Rev. A2, 1274 (1970); and M.A. Bouchiat, J. Brossel, and L.C. Pottier J. Chem. Phys. 56, 3703 (1972).
12. See, for example, M.A. Bouchiat et al., J. Chem. Phys. 56, 3703 (1972).

References

13. A.C.G. Mitchell and M.W. Zemansky, Resonance Radiation and Excited Atoms (Cambridge University Press, New York, 1971).
14. P.L. Lijnse, Fysisch Laboratorium Report 398 (Utrecht, 1972).
15. E.S. Hrycyshyn and L. Krause, Cdn. J. Phys. 48, 2761 (1978).
16. J.A. Bellisio, P. Davidovits, and P.J. Kindlmann, J. Chem. Phys. 48, 2376 (1968).
17. For recent papers, for example, J.A. Silver, C.B. Normand, and C.H. Kwei, J. Chem. Phys. 71, 3412 (1979); T.F. Gallagher, W.E. Cooke, and S.A. Edelstein, Phys. Rev. A 17, 125 (1978), and references therein.
18. F.A. Franz, R. Boggy and C.E. Sooriamoorathi, Phys. Rev. A11, 1 (1975).
19. T.R. Marshall, F.A. Franz and R. Boggy, Phys. Rev. A16, 618 (1977).
20. For a review, see, for example, W.E. Baylis, Progress in Atomic Spectroscopy, W. Hanle and H. Kleinpopper, ed., Plenum Pub. (1979).
21. J.C. Gay and W.B. Schneider, Z. fur Physik A278, 211 (1976).
22. W. Berdowski, T. Shiner, and L. Krause, Phys. Rev. A4, 984 (1971).
23. B. Niewitecka and L. Krause, Cdn. J. Phys. 53, 1499 (1975).
24. W. Berdowski and L. Krause, Phys. Rev. 165, 158 (1968).
25. J. Pascale and M-Y Perrin, J. Phys. B. 13, 1839 (1979).
26. W.E. Baylis, J. Chem. Phys. 51, 2665 (1969); revised values by private communication.

**DATE
FILMED**

2-8